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Observational constraints on the formation of Type Ia polar stratospheric clouds

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Abstract. On January 11, 1989 during AASE I, Type Ia polar stratospheric clouds (PSCs) were observed over a vast area in the Arctic. Synoptic scale temperature histories for this flight are obtained using the Goddard trajectory model. Mesoscale temperature fluctuations (MTF) are divided into two major categories of background and lee waves. The statistics of background MTFs are derived from measurements by the microwave temperature profiler aboard the ER-2 aircraft. A forecast model is used to determine the occurrence of lee waves. The MTFs are then superimposed upon the Goddard synoptic scale temperatures. Statistical correlations between temperature histories and Type Ia PSC observations indicate that MTFs can not be solely responsible for the formation of Type Ia PSCs in the stratosphere. Instead, on a synoptic scale, the exposure time of air masses to temperatures below the nitric acid trihydrate (NAT) frost point for ~ 1 day or more should be the main criteria for predicting the occurrence of Type Ia clouds in the stratosphere.

Introduction

DC-8 lidar observations during AASE I distinguished two subclasses of Type I PSCs (Type Ia and Ib) based on the scattering and depolarization behavior of the cloud particles [*Browell et al.*, 1990]. In general Type Ia particles exhibited low scattering and high depolarization ratios, whereas Type Ib PSCs showed high scattering and low depolarization ratios. Interpretations of the lidar data indicated that Type Ia PSCs are large ($r > 1$ μm) nonspherical particles, while Type Ib PSCs are small ($r \sim 0.5$ μm) spherical particles [*Toon et al.*, 1990]. However, there were observed cases of Type I PSC particles which exhibited high scattering and moderate depolarization ratios that did not agree with either Type Ia or Ib PSCs definitions [*Browell et al.*, 1990]. Such observations are more consistent with the presence of small nonspherical particles that are classed here as Type Ic PSCs [*Tabazadeh and Toon*, 1996].

The first attempt to explain why HNO_3 was condensed on both large and small size particles in adjacent air masses and at similar temperatures during AASE I, related the size of the cloud particles to the cooling rate of the air mass [*e.g.*, *Toon et al.*, 1990]. It was argued that fast cooling rates would drive higher supersaturations thereby converting more of the available sulfate cores into PSC particles, limiting their size to less than a micron. While, slow cooling rates would produce small supersaturations which would activate only a few cores into PSC particles, allowing for the formation of only large aerosols containing HNO_3. However, a drawback to this hypothesis is the fact that small mesoscale temperature fluctuations occur continuously in the stratosphere [*Gary*, 1989; *Murphy and Gary*, 1995]. This would expose all air masses to large cooling rates and convert most of the sulfate cores into some type of an initial metastable HNO_3-containing aerosol, resulting in the formation of only small PSC particles. Hence, formation of large PSC particles must involve the transfer of HNO_3 from the metastable phases to the stable phases such as NAT [*Worsnop et al.*, 1993; *Tabazadeh et al.*, 1994; Fox *et al.*, 1995; *Murphy and Gary*, 1995].

Here, we use the DC-8 observations of Type Ia particles [*Browell et al.*, 1990] and the Goddard trajectory model [*Schoeberl and Saprling*, 1995], including the statistics of background temperature fluctuations [*Gary*, 1990] and lee waves [*Bacmeister et al.*, 1994], to describe a mechanism for the formation of Type Ia PSC particles. Some recent theories suggest that the cooling of air to the ice frost point [*Koop et al.*, 1995] or the occurrence of lee waves [*Meilinger et al.*, 1995] might cause the formation of Type Ia clouds in the stratosphere from ternary solution droplets. In the ice-frost-point mechanism, Type Ia (assumed to be NAT) nucleation occurs on ice crystals formed in the solution droplets. Thus, for this processes to occur, the air temperature must drop below the ice freezing point. On the other hand, the lee wave mechanism suggest that in a rapid cooling cycle the smallest ternary droplets would freeze into NAT since their composition would be the closest to that of the trihydrate. Here we investigate whether such processes play a role in the formation of Type Ia PSCs.

Analysis Technique

To correlate the temperature experienced by an air mass with Type Ia particle observations, we follow three steps. First, the Goddard model (referred to as NMC values) is used to calculate the synoptic variations in the air mass temperature. Second, measurements from the microwave temperature profiler (MTP) aboard the ER-2 aircraft are used to derive a general Gaussian probability distribution curve for the magnitude of the background MTFs relative to the synoptic scale values [*Gary*, 1989] (In general MTFs are caused by gravity waves. For a more through description of how such fluctuations arise in the stratosphere see the work by Murphy and Gary [1995]). The Gaussian statistics are superimposed on the trajectories to investigate whether negative departures from the synoptic temperature values could reduce the air mass temperature to that of the ice frost point. This criteria is used to study the possible role of ice crystal formation in the nucleation of Type Ia clouds. Third, 24 hour lee wave forecasts are projected onto the Goddard NMC trajectory maps, going

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back for a period of 7 days [*Bacmeister et al.*, 1994]. These projected forecast maps are used to locate the regions where the air mass may have encountered a lee wave and the extent to which the actual temperatures may have varied from the synoptic values. This procedure is used to examine whether Type Ia particle observations are correlated with lee wave occurrence.

PSC Observations

Figure 1 illustrates the DC-8 flight path on January 11, 1989. Type I PSCs were observed during 5 hours of flight between points A and G marked in Figure 1. The optical characteristics of the observed PSCs and the physical interpretation of this data set is given elsewhere [*Browell et al.*, 1990; *Toon et al.*, 1990]. PSCs of Type Ia were seen at all locations except at point G. At point G, Type Ib and Ic PSCs were observed at 50 and 40 mb, respectively.

Synoptic Temperature Histories

Air mass trajectories are calculated at locations A-G marked in Figure 1 at pressure altitudes of about 40, 50 and 60 mb. The temperatures along these trajectories are shown in Figure 2. On a synoptic scale, at locations where Type Ia particles were observed (points A through F), the air temperature remained below the NAT frost point for at least a day and below the SAT (sulfuric acid tetrahydrate) melting temperature for more than a week just prior to sampling. Whereas, the air mass sampled at point G was recently cooled below the NAT frost point (trajectories G1 and G2 shown in Figure 2). Based on these observations, we infer that in order for Type Ia particles to form after temperatures drop below the NAT frost point, a nucleation time of ~ 1 day or more is required since for periods less than a day either Type Ib or Ic PSCs were observed. Our previous trajectory studies support this condition for Type Ia particle nucleation [*Tabazadeh et al.*, 1995]. We presented temperature histories of six ER-2 flights in which the observed PSCs were not composed of Type Ia PSCs (assumed to be either NAT or NAD) and all the respective air masses were cooled below the NAT frost point within the last 24 hours.

Statistics of Background Temperature Fluctuations

Murphy and Gary [1995] used the MTP data for ER-2 flights from California to Maine and calculated that for this geographic region the amplitude of background MTFs relative to the synoptic scale temperatures in the stratosphere had a Gaussian width parameter (σ) of ~ 0.8 K over water and ~ 1.6 K over land. Using MTP data for all the available Arctic flights during AASE 1 we derive a σ of ~ 0.7 K over the Arctic ocean and ~ 1.2 K over land for this region. Note that despite the geographic differences between these two regions the σs remain roughly the same.

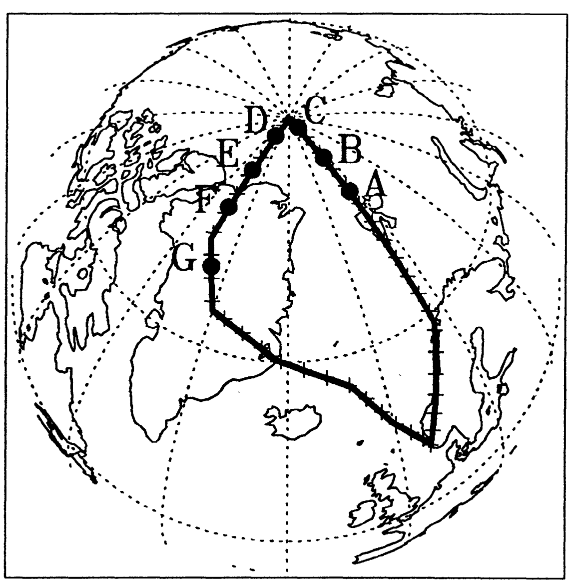

Figure 1. The DC-8 flight path on January 11, 1989. The circles indicate the locations where air mass trajectories were calculated.

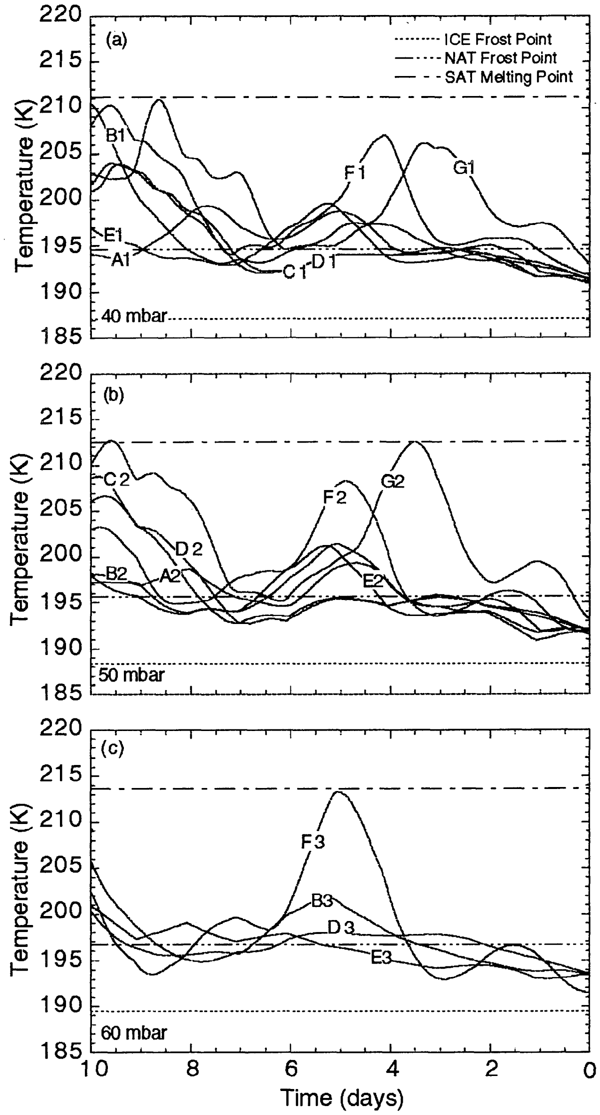

Figure 2. Temperatures on 10-day back trajectories for seven locations (marked in Figure1) where Type I PSCs were sampled by the DC-8 on January 11, 1989.

Here, we use Gaussian statistics to superimpose the effect of background MTFs upon the synoptic scale NMC temperatures. Denoting ΔT as the amplitude of fluctuation necessary to reduce the synoptic air mass temperature to that of the ice frost point, the Error Function Integral (EFI) is used to calculate the probability of how rare such a fluctuation is for a given σ (see discussions of Error Function by *Taylor* [1982]). The σs given above were calculated for ~ 6 hours of an ER-2 flight, which is equivalent to about 60 hours of an air parcel time since the ER-2 travels about 10 times faster than a typical air mass. Therefore, the statistics represented by the Gaussian distribution for an ER-2 flight correspond to the occurrence of the same fluctuations in 60 hours of an air mass time.

From statistics, if the hourly occurrence (P_1) of an event is known, then the probability for the same event to occur in t hours is given by

$$P_t = 1 - (1 - P_1)^t \qquad (1)$$

If the 60-hour probabilities (P_{60}) are known for temperature fluctuations in the stratosphere from the Gaussian distribution described above, then the hourly probabilities are related to the 60-hour probabilities by

$$P_1 = 1 - EXP\left[\frac{\ln(1-P_{60})}{60}\right] \qquad (2)$$

Substituting (2) into (1), the time dependence of the probabilities or temperature fluctuations can be calculated from the 60-hour statistics, given by

$$P_t = 1 - \left(EXP\left[\frac{\ln(1-P_{60})}{60}\right]\right)^t \qquad (3)$$

To study the role of background MTFs in lowering stratospheric temperatures to the ice frost point, we first subtracted 2 K from the NMC trajectory temperatures to allow for the maximum possible systematic error in the NMC temperatures. Figure 3 illustrates the scheme used for calculating the probability of reaching the ice frost point during the last seven days for the air mass sampled at point A (Figure 1). First, the trajectory is divided into 7 segments, lasting 24 hours each, and the minimum temperature in each segment is denoted as T_{min}. For each segment, σ was determined by using the NMC trajectory maps in order to estimate whether the air mass was over the Arctic ocean or the land mass. The amplitude of the temperature fluctuation (ΔT) needed to reach the ice frost point is calculated by subtracting T_{min} from the ice frost point temperature, assuming 5 ppmv concentration for the water vapor mixing ratio. The 60-hour probability for reaching ΔT is calculated using the EFI [*Taylor*, 1982]. The 24-hour probability is then calculated using (3). The overall probability is estimated by summing up all the 24-hour probabilities from each segment. The results for the rest of the trajectories shown in Figure 2 are summarized in Table 1.

The results shown in Table 1 indicate that it is unlikely for background MTFs to have caused the formation of Type Ia PSCs observed on January 11, 1989 through ice-cycle processing. The majority of air masses (> 80%) sampled by the DC-8 had a probability of less than 5 % to had been below the ice frost point during the last week.

Lee Wave Probabilities

The lee wave forecast model of *Bacmeister et al.* [1994] is used here to predict the occurrence of lee waves. Figures 4a and 4b illustrate the NMC calculated temperature history and trajectory map for point A (Figure 1) at 50 mb during the last week. To predict the lee wave occurrence, 24-hour lee wave forecast maps were generated using the Bacmeister et al.'s model going back for a period of seven days, starting on January 11, 1989. On each day, Figures 4a and 4b are used to estimate the air mass temperature and position, where the circle in the plots indicate the point of observation. For example, on January 11, the air mass traveled a distance between approximately the endpoint (marked as circle) and the second square from the endpoint and no lee waves were encountered on this day. Figure 4c illustrates the 24-hour lee wave forecast map on January 10, 1989. On this day, a lee wave might have been encountered upon exiting Greenland. This can be inferred by superimposing the air mass position on this day (Figure 4b) onto the lee wave map shown in Figure 4c. The possible amplitude of this lee wave event was about 6-8 K. Thus the temperature of the air parcel at the point of contact, shown in Figure 4a, could be lower than the NMC predicted temperature by up to 8 K. The results for air masses sampled at other locations are summarized in Table 1.

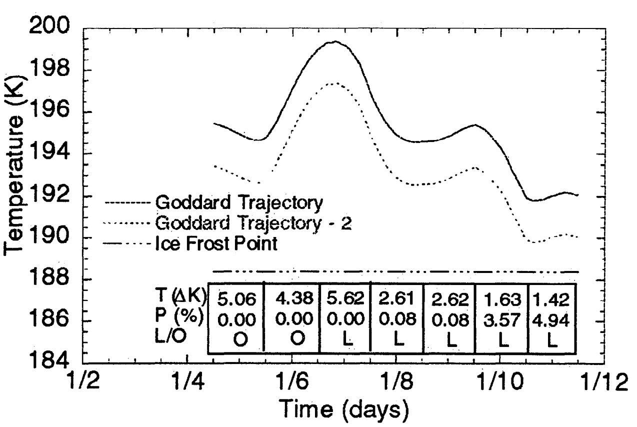

Figure 3. Temperatures on a 1-week back trajectory for point A (marked in Figure 1) at 50 mb. The scheme used to calculate the probability of reaching the ice frost point due to background temperature fluctuations is illustrated in the figure and is described in the text. Letters O and L in the plot indicate that the air mass trajectory was approximately over the Arctic ocean and the land mass, respectively.

Table 1. PSC Statistics on January 11, 1989

Location	Type	BTF (%)	LWE (day)	LWO
		40 mb		
A1	Ia	1.784	10 (6-8 K)	yes
B1	Ia	0.144	none	no
C1	Ia	0.312	none	no
D1	Ia	0.182	none	no
E1	Ia	1.763	11 (4-6 K)	yes
F1	Ia	0.490	11 (4-6 K)	yes
G1	Ic	0.052	11 (6-8 K)	---
		50 mb		
A2	Ia	8.671	10 (4-6 K)	yes
B2	Ia	3.175	none	no
C2	Ia	1.375	none	no
D2	Ia	0.590	none	no
E2	Ia	29.370	11 (4-6 K)	yes
F2	Ia	4.946	11 (4-6 K)	yes
G2	Ib	0.518	11 (6-8 K)	---
		60 mb		
B3	Ia	0.112	none	no
D3	Ia	0.052	none	no
E3	Ia	7.675	11 (4-6 K)	yes
F3	Ia	1.707	11 (4-6 K)	yes

Locations (A through G) are marked in Figure 1. The observed PSC type is indicated in column 2. The calculated probability for the background temperature fluctuations (BTF) to reach the ice frost point is given in column 3. The lee wave encounter (LWE) date and the magnitude of temperature decrease associated with this event are given in column 4. The likelihood of the lee wave occurrence (LWO) causing the formation of the observed NAT (Type Ia) cloud is given in column 5.

The results shown in Table 1 indicate that at locations in which Type Ia particles were observed only 50 % of all the sampled air masses might have experienced lee wave coolings during the last week. For example, lee waves were not encountered by air masses observed at points B through D (Figure 1) during the last week and yet Type Ia particles were seen at these locations. Specifically, at points C and D, the air mass spent the last week over the Arctic ocean, which reduces the probability of going through a lee wave cloud due to the absence of mountainous terrain. Thus if only the occurrence of lee waves leads to the formation of Type Ia clouds in the stratosphere, then at least 50 % of all the Type Ia particle observations on January 11, 1989 are unaccounted for through this mechanism.

Discussion

Based on the statistical analysis presented here, we conclude that rapid MTFs, considering both the background fluctuations and lee waves, can not be solely responsible for the formation of Type Ia particles in the stratosphere. As noted earlier, the main role that rapid fluctuations play in the

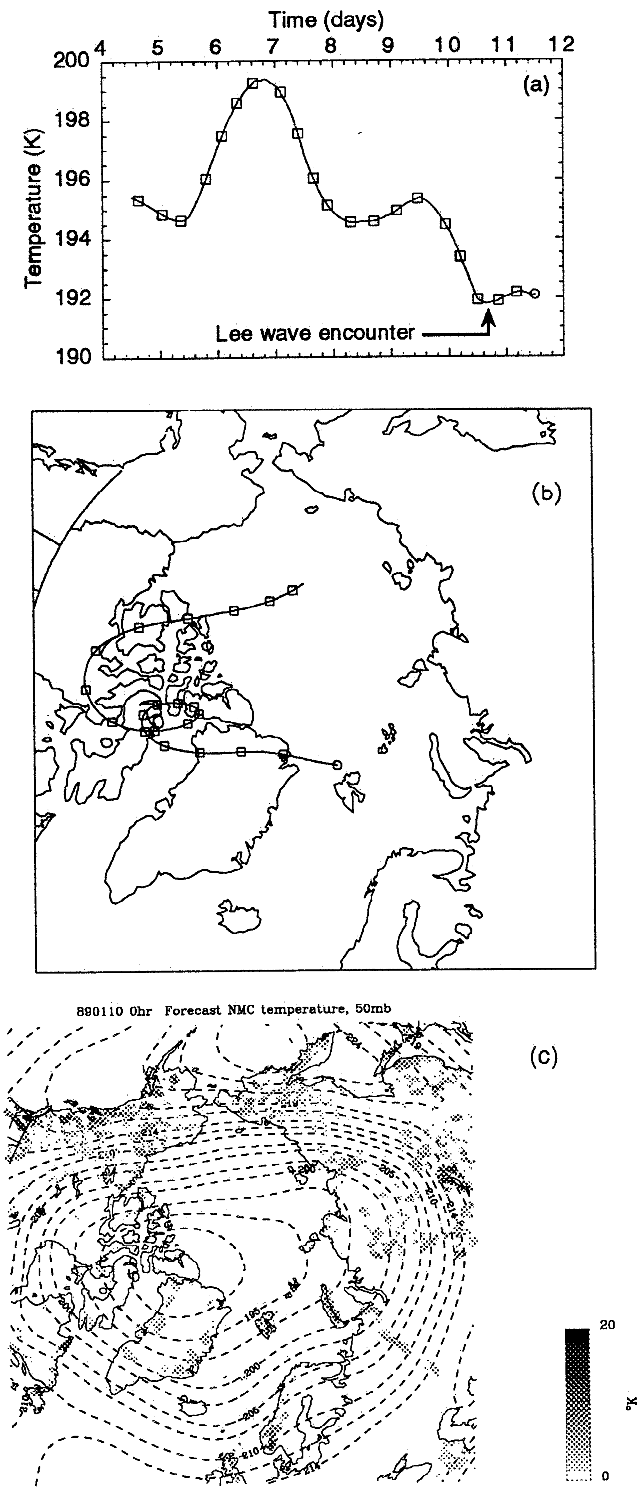

Figure 4. The NMC 1-week (a) temperature history and (b) trajectory map for point A (marked in Figure 1) at 50 mb. (c) The 24-hour lee wave forecast superimposed onto the average NMC temperatures on January 10, 1989. The relative strength of the lee wave is estimated from the scale on the right hand corner of the plot.

formation of PSC clouds is to activate most of the available sulfate cores into small HNO_3-containing particles, which are perhaps composed of metastable phases of HNO_3 and H_2O. The formation of large PSC particles must then involve the transfer of HNO_3 vapor from the metastable phases to the few particles, which are nucleated into Type Ia PSCs. According to the results discussed here, the time required for this vapor transformation to occur is approximately 1 day since for periods less than a day only metastable phases were seen by the DC-8 and similarly by the ER-2 [*Tabazadeh et al.*, 1995]. The details of the actual Type Ia nucleation mechanism from the metastable phases (Type Ib or Ic PSCs) can not be inferred from this study and requires further field, laboratory and modeling studies. Further, we have ruled out ice crystal formation or lee wave occurrence as mechanisms that are required for the formation of Type Ia clouds in the stratosphere.

Finally, we conclude that synoptic temperature values can be used to predict the occurrence of Type Ia clouds in the stratosphere. The only criteria necessary for the formation of Type Ia clouds is for synoptic temperatures to remain below the predicted NAT frost point for ~ 1 day. Similar results have been obtained recently by *Larsen et al.* [1996]. Apparently, when synoptic temperatures initially drop below about 193 K, the majority of sulfate cores grow into metastable PSC particles due to continual rapid variations in the mesoscale component of temperature and Type Ia PSCs are subsequently formed from these metastable phases. If the only condition required for Type Ia particle formation can be obtained from the synoptic temperature data, then NMC trajectory statistics can be used to predict the extent of denitrification in the stratosphere. Denitrification plays a significant role in the depletion of the ozone layer. Thus NMC temperature data can be used in future ozone related studies.

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